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Excited-Ion Lifetimes Using Penning-Ionizing Collisions*

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We show that in the Penning-ionizing collision

$$\mathrm{He}(2^3S_1)+X\rightarrow \mathrm{He}(1^1S_0)+(X^+)^*+e$$

the precession of the He^m spin at its Larmor frequency forces the J polarization of the Penning ion to rotate at the same frequency but shifted in phase. The tangent of the phase shift is proportional to the lifetime of the ionic state and presents a unique method for measuring excited-ion lifetimes.

The use of electromagnetic excitation to generate ensembles of atoms and ions in coherent states is a well-established phenomenon.[1] The coherent excitation of the system manifests itself in the Hanle effect, level crossing phenomena, and the modulation of emitted radiation. It has recently been demonstrated that coherent states can also be excited in collisions of the second kind,[2] spin-exchange collisions,[3-5] and Penning-ionizing collisions.[6] It has been suggested[6] that the latter effect will permit the application of coherence spectroscopy to states in inconvenient regions of the spectrum.

In this paper we examine the properties of the coherent states of an ion formed in a Penning-ionizing collision,

$$\mathrm{He}(2^3S_1)+X\rightarrow \mathrm{He}(1^1S_0)+(X^+)^*+e, \qquad (1)$$

between a polarized helium metastable atom and an unpolarized atom X. If the $\mathrm{He}(2^3S_1)$ atoms are prepared initially in a coherent superposition of their eigenstates by optical pumping, conservation of spin angular momentum in the Penning reaction induces coherence in the excited states of the ion. This coherence appears as a modulation of the light emitted by $(X^+)^*$ at the Larmor frequency of the He^m atoms as shown by the experiment of Schearer and Riseberg.[6]

We use the density matrix formalism to obtain the conditions under which the coherence is transferred to the ion and show that the induced coherence may be used to determine the lifetimes of excited states of the ions, particularly those which cannot be optically excited. To simplify the discussion we limit it to the ${}^2L_J$ multiplets of the ion. Conservation of the spin angular momentum in the reaction transfers the coherence of the helium metastable atoms to the ions whose transverse component of magnetization precesses at the Larmor frequency of the helium metastable state.

The state of the helium metastable atom is given by the density operator

$$\hat{\rho}_{\mathrm{He}}(t)=\tfrac{1}{4}+\tfrac{1}{2}\vec{S}(t)\cdot(\hat{\vec{S}}_1+\hat{\vec{S}}_2)+\textstyle\sum_{ij}C_{ij}(t)\hat{S}_{1i}\hat{S}_{2j}, \qquad (2)$$

$C_{ij}=C_{ji}$, $\sum_i C_{ii}=1$, and $\hat{\vec{S}}_\alpha$ is the spin operator for the "αth electron." $\vec{S}(t)$ is the average value of the J angular momentum at time t. Since the atom X is unpolarized, the initial polarization of the ion is determined entirely by the polarization of the helium metastable atom. The electron emitted in the collision comes from the helium. We then apply the spin conservation rule to the remaining electrons and obtain the initial density

operator

$$\hat{\rho}_J^{(0)} = \hat{\Lambda}_J\{[1+2\vec{S}(t_0)\cdot\hat{\vec{S}}]/(2J+1)\}\hat{\Lambda}_J,$$
$$\hat{\Lambda}_J \equiv \frac{(L+\frac{1}{2}) \pm (2\hat{\vec{L}}\cdot\hat{\vec{S}}+\frac{1}{2})}{2L+1}, \quad J = L \pm \tfrac{1}{2}, \tag{3}$$

for the ions formed in the ^2L_J multiplet at time t_0. $\hat{\Lambda}$ is the projection operator for the ^2L_J multiplet. We assume that the energy separation between the ^2L_J multiplet and other multiplets of the ion allows the neglect of those terms in the density operator coupling different multiplets. The time evolution of the state of the ion is governed by the equation

$$\partial\hat{\rho}_J/\partial t = -i[\gamma_J\vec{H}(t)\cdot\hat{\vec{J}}, \hat{\rho}_J] - \Gamma_J\hat{\rho}_J. \tag{4}$$

A common relaxation rate, Γ_J, has been taken for all matrix elements of $\hat{\rho}_J$. This should be a good approximation if the collisional relaxation of the electronic polarization is unimportant. The density operator satisfying the initial conditions and the time evolution equation has the general form

$$\hat{\rho}_J(t, t_0) = \exp[-\Gamma_J(t-t_0)]\hat{\Gamma}_J \times [1+2\alpha_J^{-1}\vec{P}(t, t_0)\cdot\hat{\vec{S}}]\hat{\Lambda}_J/(2J+1), \tag{5}$$

with

$$\alpha_J \equiv \begin{cases} L(2L+3)/3(2L+1), & J = L+\frac{1}{2}, \\ -(L+1)(2L-1)/3(2L+1), & J = L-\frac{1}{2}. \end{cases} \tag{6}$$

The density operator has been normalized such that $\mathrm{Tr}[\hat{\rho}_J(t, t_0)]$ (see the Appendix for pertinent traces) is the fraction of ions formed in the ^2L_J multiplet at time t_0 which have not decayed by time t. The time evolution of the parameter $\vec{P}(t, t_0)$ is governed by

$$d\vec{P}(t, t_0)/dt = \gamma_J\vec{H}(t)\times\vec{P}(t, t_0). \tag{7}$$

We have chosen the parameters such that $\vec{P}(t, t_0)$ is the average value of the orbital angular momentum at time t of those ions formed at time t_0. The average value of the spin angular momentum of these ions at time t is

$$\langle\hat{\vec{S}}\rangle_{t_0} = \begin{cases} P(t,t_0)/2L, & J = L+\frac{1}{2}, \\ -P(t, t_0)/(2L+1), & J = L-\frac{1}{2}. \end{cases} \tag{8}$$

The ions are formed and decay in the observation region. The observed ensemble at time t consists, therefore, of all ions in the ^2L_J multiplet at that time, i.e., the sum of the above restricted ensembles for all times t_0 prior to time t. The density operator for this system is

$$\hat{\rho}_J(t) = R_J\Gamma_J^{-1}\hat{\Lambda}_J[1+2\alpha_J^{-1}\vec{P}_0(t)\cdot\hat{\vec{S}}]\hat{\Lambda}_J/(2J+1), \tag{9}$$

where R_J is the rate of formation of ions in the ^2L_J multiplet and

$$\vec{P}_0(t) = \Gamma_J\int_{-\infty}^{t}\exp[-\Gamma(t-t_0)]\vec{P}(t, t_0)dt_0. \tag{10}$$

This density operator is normalized to the number of ions in the ^2L_J multiplet. The average orbital angular momentum for this system is $\vec{P}(t)$, while the average spin angular momentum is given by Eq. (8) with $\vec{P}(t, t_0)$ replaced by $\vec{P}(t)$. The polarization of the radiation emitted in any direction is determined by the L of the final multiplet, $\vec{P}(t)$, and conservation of angular momentum.

When the helium metastable system is prepared as described, its polarization satisfies the relationships

$$S_1(t) = iS_2(t),$$
$$S_2(t) = \chi S_0\exp(i\omega_0 t), \tag{11}$$
$$S_3(t) = S_0.$$

The J or total electronic polarization of those ions in the ^2L_J multiplets at time t is obtained from

$$p_1(t) \approx ip_2(t),$$
$$p_2(t) \approx \alpha_J\chi S_0\cos\varphi\exp[i(\omega_0 t+\varphi)], \tag{12}$$
$$p_3(t) \approx \alpha_J S_0,$$

with

$$\tan\varphi = (g_J-2)\mu_B H\tau_J, \tag{13}$$

μ_B is the Bohr magneton, g_J is the Lande factor for the particular multiplet, and τ_J is the radiative lifetime. The time dependence of the polarization is given by the angular frequency of the helium metastable state.

In the experiment performed by Schearer and Riseberg,[6] the radiation emitted perpendicular to $\vec{H}(t)$, in the transition from the $5^2P_{3/2}$ to the $5^2S_{1/2}$ levels of strontium ions, was observed. From the preceding results one can show that the intensity of this radiation is given by

$$I_\pm = \tfrac{2}{3} \pm \alpha_{3/2}S_0\cos\varphi\exp[i(\omega_0 t+\varphi)]. \tag{14}$$

The light emitted by the ion is modulated at the angular frequency of the helium metastable state, but shifted in phase. Our results are in agreement with Schearer and Riseberg's observation of the modulation of the intensity at the frequency ω_0.

To determine the lifetime of the ion, the phase

shift φ between the transverse polarizations of the helium metastable state and of the excited ionic state is measured as a function of the applied static field. Equation (13) is then used to extract the lifetime τ_J of the ionic state.

If the transverse polarization of the helium metastable atoms were stationary, the predicted effect would be the spin analog of the Hanle effect.[7] Thus, we can view the system described here as the equivalent of the Hanle effect in a reference frame rotating at the precession frequency of the helium metastable atoms.

Although the analysis of systems not satisfying the conditions imposed in this paper could be more difficult, we expect that the general properties and, in particular, the transfer of electronic polarization observed in the example would not differ substantially from those obtained here. Considering the example, we feel that Penning-ionizing collisions will prove to be an important tool in the area of ion spectroscopy.

Appendix.—In carrying out the calculations, a number of traces over products of spin and orbital angular momentum operators are encountered. It is convenient to use eigenstates of spin and orbital angular momentum rather than total angular momentum. In this case the pertinent traces decompose into products of a trace over spin states and a trace over orbital angular momentum states. The traces of particular interest are

$$\mathrm{Tr}_l \hat{L}_i = 0, \quad \mathrm{Tr}_l 1 = 2l+1,$$
$$\mathrm{Tr}_l \hat{L}_i \hat{L}_j = \tfrac{1}{3}\delta_{ij} l(l+1)(2l+1),$$
$$\mathrm{Tr}_l \hat{L}_i \hat{L}_j \hat{L}_k = \tfrac{1}{6} i\epsilon_{ijk} l(l+1)(2l+1),$$

where $l = \frac{1}{2}$ ($\hat{L}_i = \hat{S}_i$) for the spin states and $l = L$ for the orbital angular momentum states.

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Evidence for Different Interaction Potentials for He^4-He^4 and He^3-He^3 from Scattering Cross-Section Measurements

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By means of low-energy cross-section measurements on He^3-He^3, we derive an interatomic potential which has a minimum 3.4% deeper than that of He^4-He^4. We demonstrate that the difference in the potentials is significant. The atomic-mass polarization effect is proposed as a possible explanation for the different potentials.

Recently, the He^4-He^4 atomic interaction has been determined very accurately by means of scattering measurements.[1,2] It is now possible to observe mass-dependent effects in the potential, which hitherto have not been included in theoretical calculations and have not been predicted. Furthermore, the helium system is especially interesting because of the effects of quantum statistics (He^4 is a boson with spin 0, and He^3 is a fermion with spin $\frac{1}{2}$). The cross section as a function of velocity shows an undulation structure due to the statistics, and the functional dependence of the cross section versus velocity is completely different for the two systems even for identical interaction potentials. Figure 1 shows the theoretical total scattering cross sections $Q(g)$ for He^4-He^4 and He^3-He^3 calculated from potentials explained below, with application of the proper particle statistics and mass.

In this Letter we present the results of measurements of the system He^3-He^3. We have already reported measurements of the system He^4-He^4 (Ref. 1); a description of the apparatus and the evaluation procedure will be found there. Figure 2 shows a part of the He^4-He^4 measurements along with the results of the measurements